



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of Kazuo TAKAOKI

Serial No. : 10/665,574 Group Art Unit : 1713

Filed : September 22, 2003 Examiner : Rip A. Lee

For : MODIFIED PARTICLE, CATALYST COMPONENT FOR ADDITION
POLYMRIZATION, CATALYST FOR ADDITION POLYMRIZATION AND
PROCESS FOR PRODUCING ADDITION POLYMER

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DECLARATION UNDER 37 C.F.R. § 1.132

Honorable Commissioner of Patents and Trademarks
Washington, D.C. 20231

Sir:

I, Kazuo TAKAOKI, a Japanese citizen residing at 832 Pomona
Ave., #A, Albany, CA, 94706,
declare:

That I am sole inventor of the above-identified application;
That I am familiar with the prosecution history of the
identified-application; and

That the following Experiments were conducted by me or under
my direct supervision.

1. EXPERIMENT

Experiment 1 (Comparative Example)

(1) Synthesis of intermediate

Into a 2000 ml-flask, 101.2 g (0.2298 mol) of triphenylbismuth was charged, and then 500 ml of toluene was added thereto at room temperature. Thereafter, 344.8 ml of a toluene solution (2.0 mmol/ml) of pentafluorophenol (0.6896 mol) was charged, and then the obtained mixture was refluxed for 11 hours. After the refluxing, the resulted mixture was cooled to 0°C, and then a precipitated solid was collected by filtration and dried under vacuum to obtain 170.93 g of a yellow crystal.

As a result of elementary analysis, contents of Bi and F were 1.2 mmol/g and 15 mmol/g, respectively.

(2) Synthesis of catalyst component (A-1A)

In a 100 ml four necked flask purged with argon, 2.5 g (3.30 mmol) of the intermediate obtained in above (1) and 40 ml of toluene were charged. Next, 59.4 μ l (3.30 mmol) of H₂O and 3.11 g of silica (Sylopol 948, manufactured by Davison Co., Ltd.; A median particle diameter = 55 μ m; pore volume = 1.66 ml/g; specific surface area = 309 m²/g) which had been heat-treated at 300°C under a nitrogen stream, were added, then 10 ml of toluene was added thereto. The resulting mixture was heated to 80 °C and stirred for 1 hour at this temperature. After the stirring, resulted mixture was cooled to room temperature by being allowed to stand, then filtered to collect a solid. The solid was washed four times with each 30 ml of toluene, subsequently once with 30 ml of hexane, and then dried under reduced pressure at room temperature thereby obtaining 4.02g of a white powder (A-1A).

(3) Polymerization

A 0.4L-inner volume autoclave equipped with a stirrer, which had been dried under reduced pressure and thereafter purged with argon, was made vacuum, 185 ml of cyclohexane as a solvent and 15 ml of 1-hxene as a comonomer were fed therein, and then a temperature of the autoclave was raised to 180°C. Thereafter, ethylene was fed therein to obtain a partial pressure of 2.5 MPa, and the system was stabilized. To the resultant was added

0.3 mmol of triisobutylaluminum as a heptane solution, successively, 1.0 μ mol of dimethylsilylene(η^5 -tetramethylcyclopentadienyl) (3-tert-butyl-5-methyl-2-phenoxy)titanium dimethoxide and 50 mmol of triisobutylaluminum as a mixed heptane solution previously prepared, and then 69.2 mg of the component (A-1A) obtained in the above (2) were added thereto to initiate polymerization. Polymerization was carried out at 180°C for 2 minutes. As polymerization results, a trace of a polymer was trace.

Experiment 2 (Comparative Example)

(1) Synthesis of intermediate

The intermediate was synthesized in the same manner as in Experiment 1(1).

(2) Synthesis of catalyst component (A-2A)

In a 100 ml four necked flask purged with argon, 2.48 g (3.28 mmol) of $\text{Bi}(\text{OC}_6\text{F}_5)_3$ obtained in above (1) and 40 ml of toluene were charged. Next, 14.8 μ l (0.822 mmol) of H_2O and 3.06g of silica (Sylopol 948, manufactured by Davison Co., Ltd.; A median particle diameter = 55 μ m; pore volume =1.66 ml/g; specific surface area = 309 m^2/g) which had been heat-treated at 300°C

under a nitrogen stream, were added, then 10 ml of toluene was added thereto. The resulting mixture was heated to 80 °C and stirred for 1 hour at this temperature. After the stirring, resulted mixture was cooled to room temperature by being allowed to stand, then filtered to collect a solid. The solid was washed four times with each 30 ml of toluene, subsequently once with 30 ml of hexane, and then dried under reduced pressure at room temperature thereby obtaining 4.17g of a white powder (A-2A).

(3) Polymerization

Polymerization was carried out in the same manner as in Experiment 1(3) except 79.9 mg of (A-2A) obtained in (2) above was used instead of 69.6 mg of (A-1A). As polymerization results, a trace of a polymer was trace.

Experiment 3 (Comparative Example)

Polymerization

In a 400 ml-inner volume autoclave equipped with a stirrer, which had been dried under reduced pressure and thereafter purged with argon, 10 ml of 1-hexene as a comonomer and 190 ml of hexane as a solvent were charged, the reactor was heated to 70°C. After heating, ethylene was fed therein while keeping a partial pressure of 0.6 MPa. After the system was stabilized, to the resultant was added 0.25 mmol of triisobutylaluminum, successively, 0.3 μ mol of ethylenebis(indenyl)zirconium dichloride, and then 10.1 mg of the component (A-1A) obtained in Experiment 1(2) were added thereto to initiate polymerization. Polymerization was carried out at 70°C for 30 minutes.

As a polymerization result, 13.3 g of an ethylene/1-hexene copolymer was obtained. Polymerization activity was 2634

$g/\{g-(A-1A) \cdot h\}$.

Experiment 4 (Comparative Example)

Polymerization

Polymerization was carried out in the same manner as in Experiment 3 except that 13.0 mg of A-2A obtained in Experiment 2(2) instead of 10.1 mg of A-1A. As a polymerization result, 15.0 g of an ethylene/1-hexene copolymer was obtained. Polymerization activity was $2308 g/\{g-(A-2A) \cdot h\}$.

2. CONSIDERATION

(1) The results of Experiments 1 to 4, together with those of Examples 5 and 2 in the specification of the above-identified application were shown in Table 1 for comparison.

(2) It is apparent that when H₂O(water) as the component (d) in the US-2002/0143124 or DE101 64 188 A1 cited in the Office Action was used in addition to triphenylbismuth as the component (a), pentafluorophenol as the component (b) and silica as the component (c) in preparation of the catalyst component (A) and thus obtained product as the catalyst component (A) was used in polymerization of an olefin (Experiments 1 and 2, and 3 and 4), the polymerization activity of the catalysts is lower than that of the catalyst using the catalyst component (A) prepared using no water (Examples 5 and 2).

(3) These results can not be expected from the descriptions of US-2002/0143124 or DE101 64 188 A1 cited in the Office Action.

Table 1

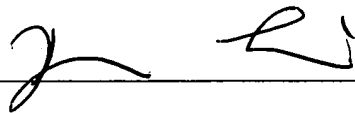
Run No.	Catalyst Component A	Catalyst component B	Catalyst component C	Polymerization	
				Polymerization result	Polymerization Activity (g-polymer/g-Cat.A/Hr)
Example 2	Intermediate/SiO ₂	Q*	Triisobutyl-aluminum		3500
Experiment 1	Intermediate/SiO ₂ /H ₂ O (Bi/H ₂ O=1/1 (molar ratio))	Q*	Triisobutyl-aluminum		2634
Experiment 2	Intermediate/SiO ₂ /H ₂ O (Bi/H ₂ O=1/0.25 (molar ratio))	Q*	Triisobutyl-aluminum		2308

Q*:Ethylenebis(indenyl)zirconium dichloride

That I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the above identified application or patent issued thereon.

Signed this

26th of April, 2005


Kazuo TAKAOKI